



Ten-year legacy of organic carbon in non-agricultural (brownfield) soils restored using green waste compost exceeds 4 per mille per annum: Benefits and trade-offs of a circular economy approach

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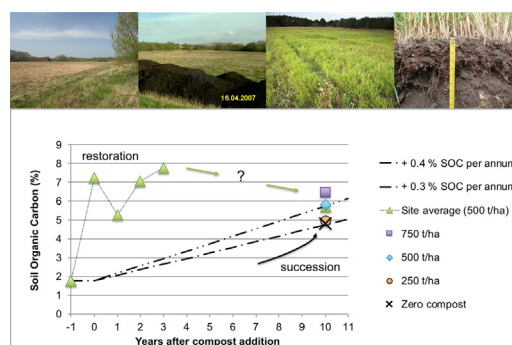
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HIGHLIGHTS

- SOC from green waste compost addition to brownfield soils declines at 7–9% pa.
- Mineralisation of N from compost is 4–5% pa under brownfield energy crops.
- Pb and Zn levels from compost decline in soil at similar rates to major nutrients.
- One-off high-rate compost applications for restoration provide >10-year benefits.
- SOC added from >500 t·ha⁻¹ compost averages 0.4% per annum after a decade.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 26 March 2019

Received in revised form 13 May 2019

Accepted 13 May 2019

Available online 16 May 2019

Editor: Frederic Coulon

Keywords:

4 per 1000

Soil carbon

Marginal land

Bioenergy

PAS100

Reclamation

ABSTRACT

Soil organic carbon (SOC) was re-analysed 10 years after application of source-segregated green waste compost at a 1 ha previously-developed UK site to compare with the increases suggested by the 4 per mille initiative proposed at COP21 in Paris. Compost prepared to PAS100 standard had been incorporated once at rates of 250, 500 and 750 t·ha⁻¹ in 2007 in clay subsoil prior to planting of perennial energy crops. Our results show statistically significant differential increases in SOC, total nutrients N and P, or contaminants Zn, Pb, Cu, As and B, remain from the compost application after a decade. For the 500 or 750 t·ha⁻¹ compost rates the SOC increments in the upper 10–15 cm were 0.85% or 1.6% over the 4.9% developed from a baseline of 1.8% in control areas by a decade of natural regeneration. Calculation of the elemental loadings from the compost analyses compared to the present-day levels suggests SOC declines after application at an average annual rate approaching 10%, compared to 5% for the nutrients or contaminants, roughly equivalent to half-lives of 5 or 10 years respectively. The study demonstrates the long-term soil organic matter (SOM) additions, fertility benefits and technical feasibility of a one-off, high-rate application of waste-derived compost to improve urban soils, compared to the potential trade-off of adding to PTE loadings. This longevity of SOC addition, previously unrecognised in brownfield soils, may be inferred for other areas where further cultivation is precluded, as is typical after landscape restoration or under perennial energy crops for the production of biomass. This unprecedented result has wider implications for marginal land use for bioenergy and the opportunities therein for SOC management using anthropogenic organic wastes to mitigate greenhouse gas emissions.

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1. Introduction

1.1. The 4 per mille initiative

The launch of the 4 per mille initiative at COP 21 in Paris in 2015 (1) was based on the observation that the annual anthropogenic atmospheric C emissions as CO₂ from fossil fuels corresponds to that fraction (i.e. 4/1000) of total global carbon stock in soils to a depth of 2 m (Minasny et al., 2017). From this observation, it is argued, increasing the existing levels of SOC in global soils by an average of 0.4% annually could provide a short-term mitigation strategy ahead of future decarbonisation, requiring a focus on improving agricultural soils with low or degraded soil conditions over the next 20 years. As the SOC of soils varies widely, so do the relative and absolute increases required, with a global C sequestration rate averaging 0.6 t·ha⁻¹ per annum. Hence the regional opportunities for implementation of this policy vary widely (Minasny et al., 2017). In the UK for example the large proportion of well-managed agricultural lands in England offer good sequestration opportunities, whereas in Scotland the soils are generally more C-rich, so action will require reductions in peatland degradation, forestry or agricultural activity simply to maintain current soil C stocks. There remain a number of outstanding scientific challenges with the political initiative from a soil science perspective (Baveye et al., 2018), including the practicalities of achieving the necessary level of storage (Minasny et al., 2017), the decline in future effectiveness of agricultural management strategies, its reversibility and or concerns regarding the longevity of C storage (Powlson et al., 2011). To achieve 4 per mille would require similar increases to be made in non-agricultural soils (Van Groenigen et al., 2017). The stoichiometric N requirements to still maintain fertility are also an obstacle, since SOM tends towards having a C:N ratio of 12:1, so SOC addition alone might even sequester plant available forms of N (Batjes, 1996; Van Groenigen et al., 2017). For example, restoration top soils should have C:N ratios < 35 and are still considered low fertility at C:N ratios ≥ 20 (British Standards Institution, 2015). Adding mineral N fertilizers to compensate would reverse any reduction of GHG emissions, due to the significant C-footprint of their manufacture or to the N₂O emissions in soils, which also exceed any soil C gains from the increased plant growth and C uptake from improved fertility (Powlson et al., 2011). As a practical consideration farmers are unlikely to have sufficient resources of farm yard manure to make the level of annual applications required to meet 4 per mille (Poulton et al., 2018). As a result the feasibility of achieving a 0.4% annual increase in SOC is still hotly debated, especially for agricultural soils (Baveye et al., 2018).

1.2. Brownfield SOC

By analogy with urban soils (Scharenbroch et al., 2005), uncultivated soils found at previously developed or potentially contaminated “brownfield” sites (Alker et al., 2000; Yount, 2003) are also likely to exhibit low nutrient status, with highly variable and often lower SOC levels than natural systems. When reused for bioenergy they can tolerate applications of organic-rich waste materials with higher C:N ratios or levels of anthropogenic contaminants than would be tolerable in agricultural systems. Long term studies suggest that repeated applications of composted urban wastes could lead to increased loadings or modified availability of various PTEs (Zn, Cu, Cr, Cd, Pb, Ni) if used in agro-systems (Cambier et al., 2019). Hence non-agricultural landbanks or “marginal” lands, such as brownfield sites, former mining sites, quarries, landfill sites, derelict underutilised neglected and peri-urban land (Lord, 2015) present an important opportunity on which to focus efforts to increase soil carbon towards meeting the 4 per mille targets. The aim of this paper is to test the hypothesis that adding green waste compost to brownfield sites during restoration or to establish energy crops could deliver long term benefits, including meeting 4 per mille SOC increases. The approach is to compare empirical observations at field trials

established over a decade earlier with baseline soil conditions, nutrient and contaminant loadings calculated for the initial compost applications.

2. Methodology

2.1. Field site

This study makes use of one of five similar 1 ha demonstration sites established as part of the BioReGen Project (Biomass, Remediation, re-Generation: Reusing brownfield sites for renewable energy crops) which was funded by the EU Life III Environment Programme (Life05 ENV/UK/000128) (2). These sites were planned using the results of a similar number of smaller pilot trials (Lord et al., 2008a, 2008b) and were designed to compare the relative productivity, contaminant uptake and fuel properties of the four energy crop species, short-rotation coppice willow (*Salix* spp.), Miscanthus (*Miscanthus* × *giganteus*), Switchgrass (*Panicum virgata*) and Reed Canarygrass (*Phalaris arundinacea*) (Lord, 2015). The Rainton Bridge trial site, near Sunderland, NE England (54.834317°N, -1.477743°E) was the site of a former sewage treatment works from at least 1896, which had been demolished prior to 1979, with the site then covered by a clay cap in 1995 and by subsoil in 2005 during preparation works for construction of the adjacent Rainton Bridge Business Park (Fig. 1). Although previously in agricultural use the area lies within the exposed part of the Durham Coalfield, with a long history of coal extraction and processing with several nearby collieries, associated wagonways, railways, brickworks and settlements. The area of the site used for the BioReGen trial had not yet been fully restored with topsoil and at the time of adoption was sparsely vegetated.

Site preparation made use of local agricultural contractors using standard agricultural equipment. At this site this involved glyphosate spraying, followed by ploughing, disking and a single compost application, the latter by surface application using a muck spreader and then incorporation of the 5, 10 or 15 cm thick layers of compost to a soil depth of c 0.1 m. The absence of layout constraints at this demonstration site allowed for the following experimental design: The roughly square 100 × 100 m trial was divided into four equal NW-SE strips of energy crops, each 25 m wide, with three zones of different rates of compost amendment arranged perpendicularly across the entire trial running NE-SW. The application rates were 750 t·ha⁻¹ (as a 25 m wide strip along NW edge), 500 t·ha⁻¹ (central 50 m wide strip) and 250 t·ha⁻¹ (25 m wide strip along SE edge). The rates chosen were multiples of the prevailing regulatory limit for annual applications to agricultural land at the time of the trial (Lord et al., 2010; Lord, 2015). Pilot studies had shown that these high rates were needed to cultivate energy crops on brownfield sites and, in particular, no area was planted without compost addition, in anticipation of total crop failure shown in earlier trials (Lord et al., 2008a, 2008b). However, the area surrounding the fenced trial was left untreated. Green waste compost prepared to PAS100 standard (Anon, 2011) was purchased from Premier Waste Management Ltd. (Coxhoe site, County Durham). This was described as “soil conditioner -fines” or the “principal grade”, so would be prepared for use as <20 mm size fraction (Eric Evans pers.com.). The compost was delivered by 20 t road wagons, stockpiled on site and then spread in zones as required using a calibrated farm loader and a muck-spreader. The application rates were all calculated for the mass of compost as received, which had a dry matter content of 61–70%. Refrigerated representative samples were taken for analysis from the stockpile of compost delivered to this site and those at other sites (Table S1). Compost application was followed by repeated disking in an attempt to mix the compost with the surface soil, prior to planting, pesticide spraying and rolling. Shortly after planting of the cuttings, rhizomes and seeds the site became waterlogged, following prolonged torrential rain, with 219 mm falling in June and July 2007, approximately double the 30-year average (3). Of the five demonstration sites planted in the BioReGen Project, that at Rainton Bridge showed the



Fig. 1. Layout of trial and compost application rates at the Rainton Bridge site near Houghton-le-Spring, Sunderland UK (54.834317°N, -1.477743°E).

strongest overall growth over the first 3 seasons, especially for reed canarygrass. Other publications give details of the practical methodologies (Lord et al., 2008b), use of compost (Lord et al., 2010), interim (Lord and Green, 2011) and final results (Lord, 2015) of the energy crop trials up to 5 years after establishment. No further cultivation or compost application has occurred at the site since establishment, other than the mechanical harvesting of reed canarygrass and manual cutting and collection of willows and *Miscanthus* after the 2008 and 2009 growth seasons.

2.2. Soil sampling protocols

In this study, representative new samples were obtained to characterise the SOC and nutrient status of soils in each compost application rate plot and across an unamended control area. Four replicate composite samples were collected using a 2.5 cm diameter gouge auger (Eijkelpkamp), collecting single cores at 25 equally-spaced subsamples sites in a W-shaped traverse crossing each rectangular plot. In each case the five apices of the four sectors of the traverse were first marked out with bamboo canes, then the intervening five sampling locations on each sector paced out and marked by survey poles. Darkened soils representing the depth of compost incorporation were not observed to extend to >0.15 m below the surface in the 750 t·ha⁻¹ plot where the most compost was used, so this sampling depth was adopted throughout, marking the auger accordingly. The replicate samples were collected from each plot, by treating each of the four sides in turn as the top of the W-shaped traverse, then spacing the sample collection points to cross the length and width of the plot. Control samples were collected in four similar traverses in a 25 m wide area parallel to the WNW margin of the trial where no compost had been spread (Fig. 1). This area includes the access route to the planting trial for agricultural equipment but otherwise had been left to regenerate naturally after establishment and fencing work.

The soil cores from each traverse were homogenised in the field using a stainless-steel trowel and a plastic bucket, prior to refrigeration and despatch to the laboratory by courier.

Soil samples were also collected for characterisation and verification purposes during the original compost addition trial and during monitoring of the energy crop field trial. As some of these required different protocols, these are reviewed here to allow for valid comparison of the original findings with data for the current study (Table S2). Three baseline samples were collected in 2006 to determine the level of contamination and nutrient status of surface soils in locations spanning the likely trial area ahead of implementation. These were collected as single “grab” samples using a stainless steel border spade to a depth of 0.1 m and area of 0.2 m × 0.2 m, with removal of vegetation and field homogenisation (Lord, 2015). This method was adopted partly to sample the available growth medium and partly to avoid damaging the technical capping present here. Single field-homogenised samples were used originally, rather than bulking several subsamples to make a composite sample, since spatial composite sampling should not normally be used for the investigation of land potentially affected by contamination (British Standard Institution, 2017). The first phase of site monitoring commenced in summer 2007 immediately following site preparation, compost application and planting. The same sample collection protocol was used for this as in the baseline sampling but with soils collected at the centre of the site and c 10 m away from either fence inside each corner of the enclosure. These five locations represented different combinations of compost rate and plants as follows: (i) Short-rotation coppice (SRC) willow with 750 t·ha⁻¹ compost, (ii) SRC willow and 250 t·ha⁻¹, (iii) switchgrass and 250 t·ha⁻¹, (iv) switchgrass and 750 t·ha⁻¹, (v) centrally, on the dividing line between *Miscanthus*/reed canarygrass plots, both with 500 t·ha⁻¹. Consequently, the mean compost addition rate is 500 t·ha⁻¹ for each annual set of samples, the

average for the site as a whole, whereas the range includes any variation between $250 \text{ t} \cdot \text{ha}^{-1}$ and $750 \text{ t} \cdot \text{ha}^{-1}$. Individual analyses for each sample site and monitoring campaign are given in the linked data. Sampling was repeated for three subsequent years (2008–10) at the same locations in an attempt to monitor changes in the effects of the compost addition. In the final year of monitoring, duplicate 0.1 m cores were collected at 25 subsites along W-shaped traverses across each of the compost application rates in the area of one plant species (RCG), since this had grown prolifically at this site in the areas with more compost (Lord and Green, 2011). Averages and ranges for all samples in each monitoring year and or sampling method are shown in Table S2.

2.3. Analysis

The same commercial laboratory (NRM Laboratories) was used for analysis of samples from all sampling campaigns, with the same suites of parameters. The 59 determinants on each sample included a range of inorganic and organic contaminants (including PTEs, total petroleum hydrocarbons, phenols and speciated PAHs), physio-chemical parameters (including pH, conductivity, moisture content, sand/silt/clay textural class), total and available nutrients (N, P, K, S, Ca, Mg). The analytical protocols and internal procedures employed by the laboratory are based on standard UK (or US) methods for soils and wastes (e.g. (Jackson et al., 1986)) for determining agricultural parameters specified in the relevant guidance (e.g. Anon, 2019; British Standards Institution, 2017). For conciseness, only the analytical methods for those determinants which gave significantly different results for the different compost application rate plots are described here in detail.

In the baseline and initial monitoring analysis SOM was reported by the laboratory, determined using the Walkley-Black (Walkley and Black, 1934) wet oxidation method. In this method potassium dichromate and then strong sulphuric acid are added to the soil, with SOC determined by its partial reduction of the former without additional heating, so any C present as carbonate is lost on acidification and is not reported. Calculations assume that 75% of SOC is reduced and that SOM contains 58% SOC. This method is potentially advantageous for determining the fertility of brownfield soils in industrial coalfield regions in that geogenic or pyrogenic C in coal or black carbon are known to be more resistant to wet oxidation than recent SOM forms (Chan et al., 2017; Siavalas et al., 2013). For the present campaign, however, the laboratory had withdrawn this method to avoid the use of potassium dichromate. Instead a Dumas method based on oxygen-rich combustion at 1000°C was used on soil dried at 30°C and finely ground in a vibratory disc mill (British Standards Institution, 2012). The Dumas method returns total carbon so either the sample must be pre-treated with acid to remove carbonate, as in this case, or inorganic carbon (IC) is determined separately and deducted (4). In both cases SOM is then estimated by applying the factor 1.724 (Pribyl, 2010) to the SOC content. Pre-acidification suffers from incomplete removal of some unreactive carbonates (e.g. dolomite), while clays or some forms of organic matter may be lost by oxidation (Hoogsteen et al., 2018). As only SOM was originally reported for the wet oxidation analyses, this has been back-calculated to SOC here as the most suitable common parameter for comparison purposes. In contrast, the SOM content of compost samples was determined directly by loss on ignition (LOI), as is typical for the characterisation of organic wastes, which is performed at a sufficiently low temperature (430°C) to avoid decomposition of any carbonate that might be present. The SOC of compost was then estimated by dividing LOI by the same conversion factor as is used for soils.

Observations during sieving of newly collected replicate soil samples had shown the presence of discrete particles of Magnesian Limestone, coal, coke or breeze, plastic and humic material, possibly derived from compost, as well as recent biomass as sections of grass stem or roots. To provide suitable confirmatory information on total organic carbon (TOC) contents and analysis of IC contents for this variety of C phases in a dolomitic clay soil, all current soil samples were re-analysed in

triplicate by sequential LOI at 550°C and at 950°C respectively at the University of Strathclyde using the following method: Air-dried $<2 \text{ mm}$ soil fractions were cone-quartered and c. 50 g was ground in an agate ball mill (Retsch PM100) to homogenise and reduce particulate organic matter. Consistent target masses of 1 g were dried overnight at 105°C in pre-combusted porcelain crucibles prior to ashing at each temperature in turn (Nabatherm P330) for 4 h, allowing to cool to ambient temperature in a desiccator prior to weighing to the nearest 0.1 mg after each combustion. The combination of combustion temperatures and times were chosen for the following reasons: Although LOI is routinely used to determine SOC (Nayak et al., 2019), factors including sample size, exposure time and furnace position can all effect the results unless standardised, especially at 550°C (Heiri et al., 2001). Lower temperatures are often used for SOC determination to avoid overestimates due to loss of structural water from clays or partial oxidation of certain carbonates (e.g. siderite) from 500°C (Hoogsteen et al., 2018; Pallasser et al., 2013). Conversely, this may prevent full oxidation of SOC (Nayak et al., 2019), particularly for shorter ashing times (Heiri et al., 2001) or when thermally resistant C forms or coal are present (Siavalas et al., 2013). Since LOI at 550°C could potentially lead to an overestimated SOC content in clay soils containing coal, coke, plastics or black carbon, it is henceforth denoted as $\text{TOC}_{500^\circ\text{C}}$, whereas by consensus any subsequent LOI between 500°C and 950°C can be wholly attributed to decomposition of carbonates including dolomite or calcite, from which $\text{IC}_{500-950^\circ\text{C}}$ a minimum value of IC, can then be calculated.

Total N was also determined by a Dumas method using an air-dried ($<30^\circ\text{C}$) sample sieved or ground to pass a 0.5 mm screen (Helrich, 1990). Hot water extractable B from an air-dried ($<30^\circ\text{C}$) sample sieved to pass a 2 mm screen was determined by Inductively Coupled Plasma Optical Spectroscopy (ICP-OES) (Jackson et al., 1986). Soils dried at 30°C and ground to pass a 0.5 mm screen are digested in hot aqua-regia to give “total” concentrations of elements, including total P, K, Ca, Mg, Pb, Zn and As by ICP-OES or Inductively Coupled Plasma Mass Spectrometry (EPA, 1996; Jackson et al., 1986).

2.4. Data quality and processing

Due to the likely importance of SOC measurements in the present study, all samples submitted from the 2010–11 sampling campaign were analysed in duplicate by the same laboratory for this determinant. Statistical analysis was performed using IBM SPSS Statistics version 25, including a one-way multivariate analysis of variance (MANOVA) to compare the composition of soils from different compost amendment plots to see if the differences were significant. When data from the initial batch of 16 samples collected on 22–25 September 2017 were examined it became apparent that the first and second samples taken from the $750 \text{ t} \cdot \text{ha}^{-1}$ plot had returned the lowest (4.1%) and then the highest (7.2%) overall concentrations in the entire survey, whereas other sample groups were more reproducible and roughly proportional to application rates. Re-analysis of the same $<2 \text{ mm}$ laboratory samples gave similar results, with the reported concentrations of SOC showing a positive correlation between replicates which was significant at the 99% level (Pearson correlation coefficient = 0.901, two-tailed test $p < 0.01$, Fig. 2a). Since these soils were both collected as 15 cm cores from 25 subsample locations in mirror-imaged W-shaped traverses over the same $100 \text{ m} \times 25 \text{ m}$ area, this internal discrepancy between replicates was very difficult to explain, other than as the result of either poor field homogenisation or laboratory subsampling. The samples were observed in the field to consist of a friable compost-rich soil horizon lying above a stiff grey clay (Fig. 3), rendering field homogenisation of the 25 cores very difficult, and unrepresentative laboratory subsampling of a heterogeneous field sample a distinct possibility. Initial sieving to $<2 \text{ mm}$ must be performed without drying if nutrients are to be determined, without the opportunity for drying and disaggregation, for which this laboratory used a shatter box. Furthermore, the samples from this brownfield site were seen to include carbon in a number of

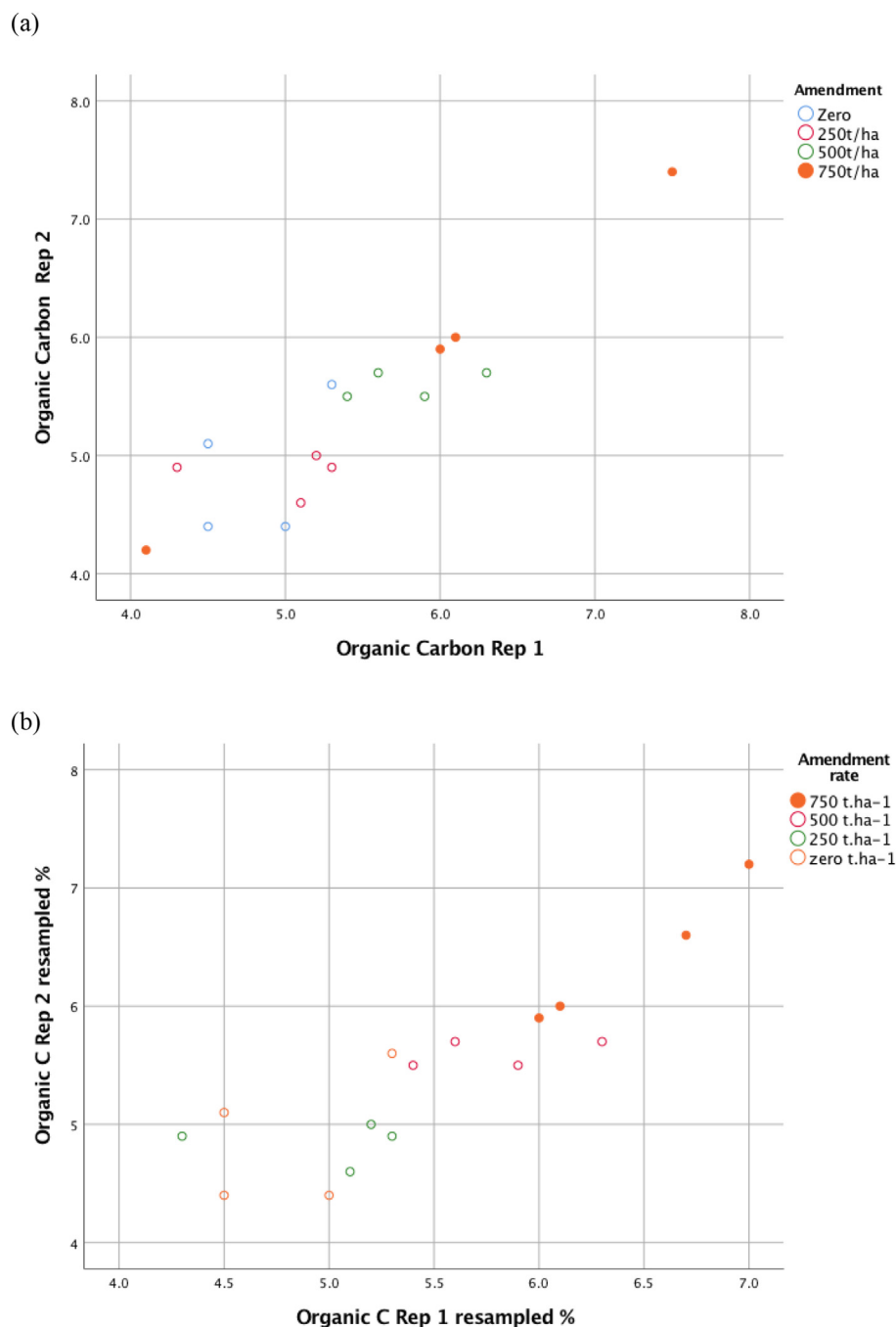


Fig. 2. Comparison of replicate analyses for SOC in compost amended areas after 10 years (a) initial samples including anomalously high and low outliers for 750 t·ha⁻¹ ($n = 16$, Pearson correlation $r^2 = 0.901$, >99% significance), and (b) after resampling and analysis of outliers ($n = 16$, Pearson correlation $r^2 = 0.881$, >99% significance).

forms, so it is possible that conventional laboratory subsampling procedures designed for typical agricultural soils were not entirely suitable for the bipartite, complex and highly variable technosol soil matrix found here. The sample with the highest SOC content also showed anomalously high levels of Zn, Pb, Cu, As, an elemental association found in the compost, which would support the hypothesis that the internal variation between sample from the compost-rich area reflects preferential subsampling of this component. Eventually it was decided to recollect both anomalous samples, which due to seasonal weather was then delayed until 10 March 2018. This eliminated the anomalously low result in the first sample, produced a reasonable grouping (Fig. 2b) and again a significant correlation between replicate analyses (Pearson correlation coefficient = 0.881, two-tailed test $p < 0.01$). For completeness, Table S2 summarizes all data from both the original and resampled

analyses, since the outliers in the former set slightly extend the overall range of reported values for the site as a whole. Thereafter, when data for different compost application rates are separated (Tables S2–S4, Fig. 4), only the later substituted analyses are included in the groups, representing “cleaned” data with outliers removed.

3. Results

3.1. Compost analysis

The set of samples from each trial site stockpile (Table S1) can be used to infer the degree of internal variation or consistency to be expected in the compost, as applied to the trial site under investigation. The majority of parameters show acceptably low variability, with



Fig. 3. Photograph of 0.15 m soil cores from gouge auger subsampling prior to field homogenisation, showing downward decrease in SOM and increase in grey to yellow clay (e.g. lower core). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

relative standard deviations below 25%. The exceptions (in bold for >25% RSD, bold italic for >50% RSD) for contaminants are Ni, Cr, sulphide, benzo(a)pyrene and the totals for all PAHs, which are all present at low concentrations and each PTE is well below the threshold for PAS100 compost. The contaminants Zn, Pb and Cu occur at higher absolute concentrations, albeit still well below the threshold values in this case. Variation in nutrients is most pronounced for the two available N species that were analysed, nitrate and ammonium, both of which show outliers and are at trace levels. In terms of the other major nutrients present in the compost these are ranked Ca, Mg or K, then P or S in order of decreasing total concentration. The high total concentrations of Ca and Mg presumably reflect the manufacture in a former Magnesian Limestone quarry in Coxhoe, Co. Durham and green waste provenance from the surrounding area. The proportion of available K in the compost (55%) is noticeably higher than for either N (2.7%) or P (5.6%).

3.2. Soil analysis during energy crop trial (years -1 to +3)

Table S2 summarizes the results of all soil sample monitoring before, during, annually afterwards and then in this current campaign > 10 years after compost addition. Most parameters show a spread of values, partly as the earlier monitoring sample set locations included areas with each of the levels of compost application and partly because the incorporation proved to be heterogeneous at the scale of a 1 kg sample. However, the following generalised observations can be drawn: Firstly, compost addition has an immediate beneficial effect on the mechanical and textural properties. Particle size analysis shows an increase in the proportion of 2–0.06 mm (sand) size fraction at the expense of the <0.002 (clay) size fraction, presumably due to the coarser dominant size fraction of the compost. As the LOI from organic matter in compost is $32 \pm 5\%$, so the inorganic component must be $68 \pm 5\%$, presumably from sandy soil included in the garden waste feedstock to the

composting process. This leads to a different particle size class using the UK Soil Survey of England and Wales textural classification scheme (Avery, 1990, 1973), now falling within the clay loam field rather than that for clay. This shift in the apparent textural soil type persists for over 10 years. The compost typically leads to a slight decrease in soil density and in a lower dry solids content (and hence presumably a greater water content at field capacity). Any reduction in pH and increases in conductivity from compost addition are small. In contrast, average SOM content, N_T , P_T and K_T concentrations increase by factors of between three and four after compost addition (Table S2). Increases in the available concentrations of P and K are even larger, increased by factors of 5–10.

In all cases these step changes persist in future years, at least over the initial three-year monitoring period. Of the major nutrients, K shows the most obvious systematic decline in future years, especially for K_A . The compost has a slightly higher Ca/Mg (2.3 atomic ratio) compared to the receiving soils (1.3), which raises the ratio of the amended soils (1.6, average for 2007–2010) but has no obvious effect on the total or available concentrations of either element in the amended soils initially. While the long-term ratio and availability of either element is little changed, the total concentrations of Ca and Mg appear to increase steadily, presumably due to the reduction in dilution by SOM. Disadvantageous changes due to compost-born contaminants (see limits in Table S1) are rarely at levels of concern for phytotoxicity, other than possibly for B (Table S2), which might approach significant levels (Nable et al., 1997) in compost before incorporation, as has been discussed elsewhere (Lord and Green, 2011; Lord, 2015). Other PTEs or anthropogenic hydrocarbons show small increases (e.g. for Cd, Hg, Pb, Zn, total petroleum or PAH) and are well below levels of regulatory concern.

3.3. Longer term soil effects (years +10 to +11)

The legacy of compost addition at different application rates on the residual soil compositions are compared both statistically (Table S3) and graphically (Fig. 4) for total concentrations of the five major nutrients, the main PTEs observed, SOC, $TOC_{550^\circ C}$ and $IC_{550-950^\circ C}$. Of these, P_T , Zn, N_T , $TOC_{550^\circ C}$, B, SOC, Pb, As and $IC_{550-950^\circ C}$ in rank order of Fisher coefficient, gave significant differences between the means for the different compost rates, in most cases between the 750 t·ha⁻¹ or 500 t·ha⁻¹ plots and the zero or 250 t·ha⁻¹ pairs. Compared to the baseline, clear increases in these parameters are still seen in the average soil concentrations after ten years, roughly in proportion to the amount of compost added. The average SOC contents after 10 years (Table S4) of 6.44% (750 t·ha⁻¹) or 5.70% (500 t·ha⁻¹) are respectively 1.59% or 0.85% above the 4.85% found in the unamended soils (0 t·ha⁻¹), equivalent to relative increases of 32.7% and 17.5% compared to no compost addition or cultivation. When comparisons are made to the average SOC (1.76%) of the three baseline grab samples collected before the trial began these increases are even more extreme at 4.68% and 3.94%, relative increases of 266% and 224%, although with the minor caveat that the sampling methods were different. A similar rise can be seen for several other determinants, including K_T , but this difference is far more pronounced for SOC. This reflects the much greater difference between the SOC content in the unamended soils of the energy crop trials after 10 years and the subsoil conditions before the trial began, whereas for most other determinants they are more similar (Table S4). Curiously, concentrations of $IC_{550-950^\circ C}$, Ca and Mg show the reverse pattern to $TOC_{550^\circ C}$ and fall with increasing compost rate and residual SOC. Presumably this reflects greater residual dilution of the dolomite-bearing clay soil by organic matter, as deduced from the observed progressive rise in Ca and Mg over time. Although K_T shows a pronounced step change on application (Table S2), after 10 years there appears to be a fairly homogenous distribution through the site with no significant correlation with the amount of compost added

(Fig. 4, Table S4). Likewise, any changes in Ca, Mg or Ca/Mg ratio in the more highly amended areas are not statistically significant.

3.4. Nutrient release and contaminant fate (years +10 to +11)

Using the average composition of compost at delivery, density, moisture content (Table S1) and as received application rates ($750 \text{ t} \cdot \text{ha}^{-1}$, $500 \text{ t} \cdot \text{ha}^{-1}$, $250 \text{ t} \cdot \text{ha}^{-1}$) the likely mass of each nutrient and contaminant added to the soil can be estimated for each area (Table S5a). These have been compared to the residual loadings present in the amended plots after 10 years (Table S5b), from which the differences in concentrations compared to those in the unamended soils are calculated (Table S5c). Approximately 50–60% of the N_T , P_T , Zn, Pb and Cu etc. appear to have been retained in the soils where $750 \text{ t} \cdot \text{ha}^{-1}$ or $500 \text{ t} \cdot \text{ha}^{-1}$ of compost was added, compared to about 20–25% of the SOC (Table S5c). For N and P this equates to an annual release rate of 4–5%, compared to 9–10% for K (Table S6). For N at the highest compost application rate

this is equivalent to a loss of $237 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{a}^{-1}$, compared to a lower original leachable content of $128 \text{ kg} \cdot \text{ha}^{-1}$ on application, presumably released by catabolism and mineralisation. Both are markedly lower than the $4719 \text{ kg} \cdot \text{ha}^{-1}$ total N that the waste-derived compost addition contained. For the highest compost rate K_T declines by $292 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{a}^{-1}$, equivalent to $9.2\% \cdot \text{a}^{-1}$ and up to $9.8\% \cdot \text{a}^{-1}$ at lower rates, so has very nearly returned to uniform concentrations after a decade, albeit at a higher level than before amendment (Fig. 4). The fate of this K is unknown, as only a fraction of it can be accounted for by energy crop offtake during the original energy crop trial, even when the relatively high content of this element in the *Miscanthus* and *Phalaris* grown is considered (Lord, 2015).

Before the trial began the average SOM content was determined as 3.0%. Dividing by the minimum consensus conversion factor ($\text{OM} = 1.724 \text{ OC}$) (Pribyl, 2010) gives an upper estimate of 1.76% for SOC in 2006–7, before the trial began. A modelled annual compound increase of 4 per mille from this baseline figure for 10 years gives an SOC target

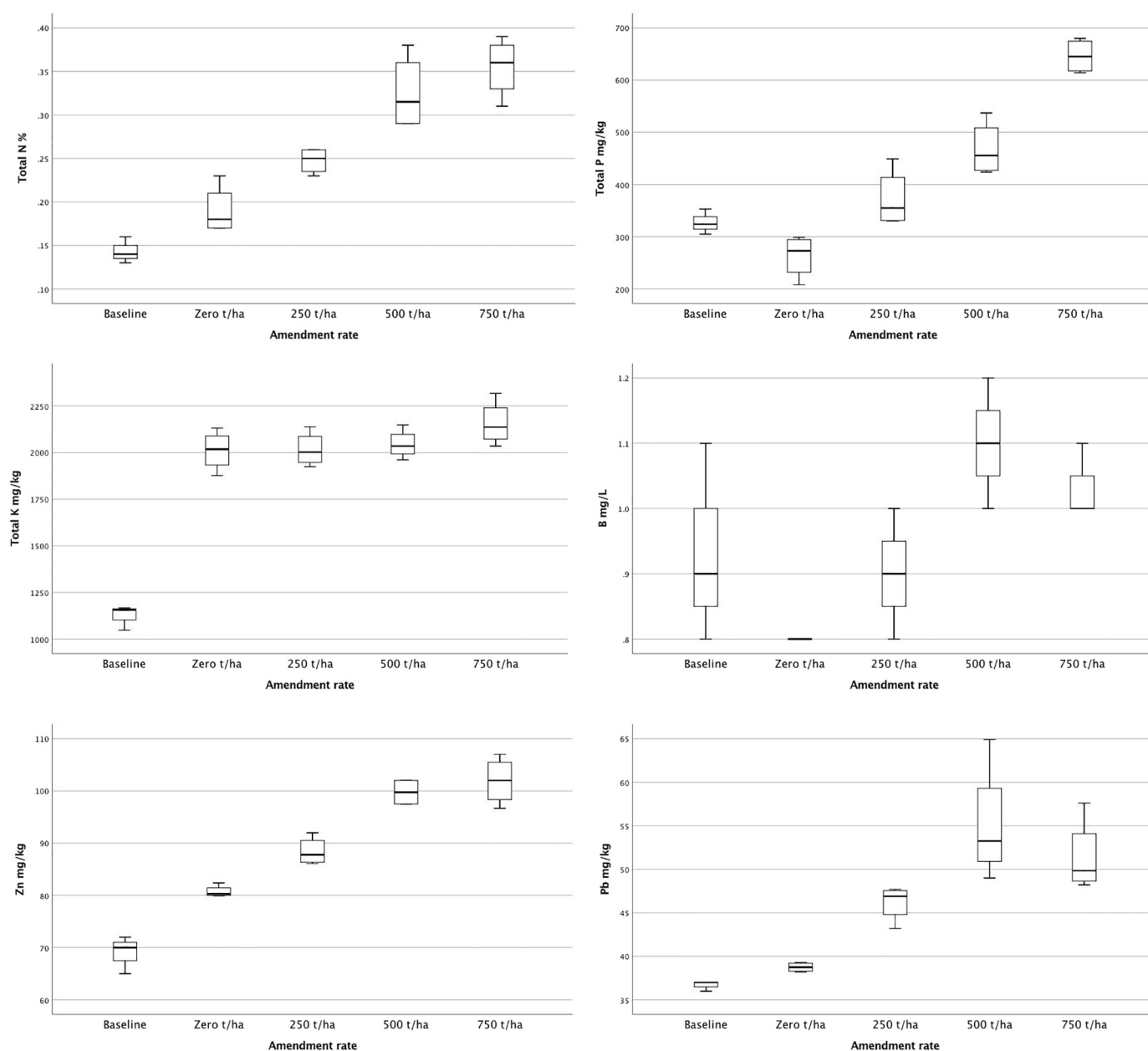


Fig. 4. Selected box plots comparing analyses of soils from each compost application rate ($n = 4$) after ten years and baseline ($n = 3$) before amendment where available.

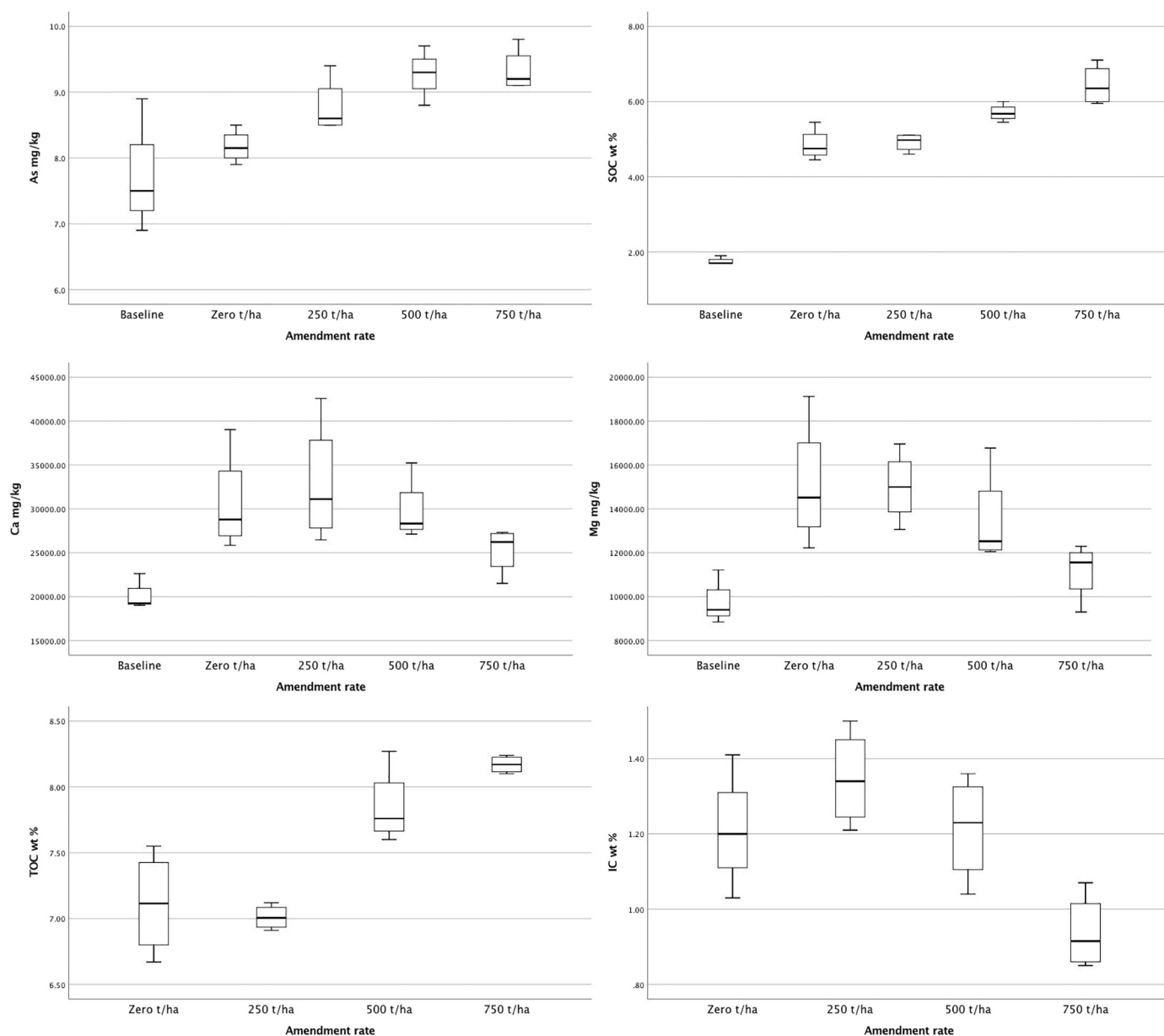


Fig. 4 (continued).

figure of only 1.83% for 2017 (Fig. 5). Growth at 4% with the same model conditions gives an SOC target of 2.60%. If the global average annual increase of $0.6 \text{ t} \cdot \text{ha}^{-1} \text{C}$ required to achieve 4 per mille (Minasny et al., 2017) is expressed as an increase in SOC concentration only within the upper 0.15 m, this falls between the 4 per mille and 4% growth models, reaching 2.19% in 2017. In comparison our field trial results show on average over the ten-year period an absolute increase of c.0.4% SOC per annum is required to match those levels found in the upper 0.15 m of soils where a single addition of $500 \text{ t} \cdot \text{ha}^{-1}$ or more compost was applied (Fig. 5). The 0.4% SOC per annum model gives 5.76% SOC by 2017, equivalent to adding $5.6 \text{ t} \cdot \text{ha}^{-1} \cdot \text{a}^{-1} \text{C}$, thus exceeding the global average figure of $0.6 \text{ t} \cdot \text{ha}^{-1}$ by a factor >9. However, when the unamended and unplanted control areas which were allowed to naturally regenerate are also compared to the baseline analyses before the trial, these also show levels exceeding those after absolute increases of 0.3% SOC per annum over the upper 0.15 m of topsoil, or 4.76% SOC and a factor of 7 over the global average.

4. Discussion

4.1. Opportunities for use of organic wastes in land restoration and soil C storage

Introduction of the EU Landfill Directive 1999/31/EC (Council Directive, 1999) and associated policy measures, such as the UK Landfill Tax in 1996 and its escalation since 1999, have encouraged the steadily increasing diversion of biodegradable municipal wastes from landfill disposal. The growing availability of recycled organic materials, such as those meeting the Publicly Available Standards (PAS100) source-segregated green waste compost (Anon, 2011) or PAS110 anaerobic digestate fibre (Anon, 2010), has enabled supply and increased the cost-effective use of these and other organic wastes in land restoration. From a regulatory perspective, our results confirm that this can be done using such material without significant risk of harm to the environment from the PTEs or total nutrient loads likely to be present in compost

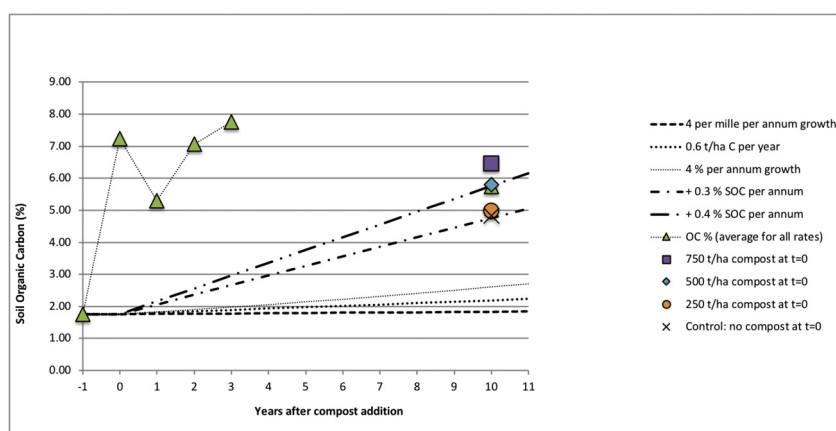


Fig. 5. Initial and long-term SOC content of brownfield soils following PAS100 compost addition. Symbols show actual data from analyses whereas dashed lines show calculated trends. The various trajectories are modelled as annual increases from the baseline SOC from the year of compost addition onwards. 4 per mille and 4% growth in the indigenous soil SOC are compared to the $0.6 \text{ t} \cdot \text{ha}^{-1}$ global average requirement for 4 per mille (Minasny et al., 2017), together with 0.3% and 0.4% annual additions of SOC needed to match the net increases that are observed here.

(Table S1). During implementation of this field trial both land restoration and soft landscaping options were introduced as designated market sectors in the Quality Protocol for compost (Environment Agency, 2012), allowing quality compost prepared to the PAS100 standard to be used as a recovered product, rather than a waste material subject to regulatory controls (Lord et al., 2010). At that time the Waste Resources Action Programme estimated the UK production of compost to be over $2 \text{ Mt} \cdot \text{a}^{-1}$ of which 17% was used in landscaping and land restoration (WRAP, 2008). Funding for demonstration trials to support development of these markets led to a number of successful field scale demonstrations as “trailblazer” projects across the UK (Edwards et al., 2012; Mathers, 2008), including the original establishment of the site used here. In comparison in 2009 the last complete National Land Use Database survey recorded nearly 69,000 ha of previously-developed land in England (Homes and Communities Agency, 2010). To these brownfields a number of other non-agricultural or “marginal” land types can be added where compost might be used for restoration, including former landfills, abandoned mine lands, quarries, contaminated or degraded land, derelict underutilised or neglected peri-urban land (Lord, 2015). Our recent research shows that at a city or country scale the inclusion of landfills and abandoned mine lands alone can more than double the potential landbank (Donaldson and Lord, 2018; Mellor, 2018). The societal and environmental benefits of this style of land reuse and recycling of urban waste align with UN Sustainable Development Goals 11, 12, 13 and 15. When combined with energy crops or other land-based renewables as part of the “energyscape” then a variety of other ecosystem services may be delivered alongside soil carbon regulation (Burgess et al., 2012; Coleby et al., 2012; Howard et al., 2013). The conclusion is that a significant opportunity exists to use compost in restoration activities of the type investigated here, or on marginal land, in the UK and worldwide, provided the benefits and trade-offs are fully understood.

4.2. Short-term benefits and trade-offs of compost addition to soil

The results of our field study largely confirm the immediate benefits of adding waste compost to soils during restoration to enhance plant growth, as are comprehensively described in academic and technical guidance (Bending et al., 1999; Claassen and Carey, 2004; Cogger, 2005; Edwards et al., 2012; Nason et al., 2007; SNIFER, 2010), albeit for a wide variety of input materials with variable compositions. For real applications the selection of organic amendment material is often determined simply by local availability or price. However, a growing number of case studies support the use of source-segregated green waste compost in a variety of restoration contexts from regeneration

of brownfield land (CL:AIRE, 2008; Kilbride, 2014), greenspace creation (Sæbø and Ferrini, 2006), reuse of contaminated or remediated soils (Rodriguez et al., 2010) manufactured soils (Palmer and Davies, 2014) or energy crop establishment (Bardos et al., 2011; Paulson et al., 2003). In the early stages this trial showed the benefits of surface incorporation of compost, with a single preparatory application rate of $500 \text{ t} \cdot \text{ha}^{-1}$ providing the best trade-off between early years establishment and future yield (Lord et al., 2010; Lord and Green, 2011; Lord, 2015).

A previous incubation study used soils taken from this site in May 2009 in the control area and three locations where $750 \text{ t} \cdot \text{ha}^{-1}$ had been incorporated in 2007 (Whelan, 2011; Whelan et al., 2013). The results showed limited additional available nutrient benefit from either adding $>500 \text{ t} \cdot \text{ha}^{-1}$ compost or from incremental compost additions at 250, 500 or $750 \text{ t} \cdot \text{ha}^{-1}$ respectively, particularly for total oxides of N, which were considered to be a limiting nutrient for plant growth (Whelan, 2011). Measurements of hydraulic properties and physical quality on the four field soils indicated that compost addition improved properties such as water holding capacity, macroporosity and hydraulic conductivity but the effects of adding $>500 \text{ t} \cdot \text{ha}^{-1}$ compost were limited. Furthermore, numerical simulations of the soil profiles exposed by trial pits in July 2010 using Hydrus-1D suggested that the $250 \text{ t} \cdot \text{ha}^{-1}$ soil profile was often too dry, whereas the $750 \text{ t} \cdot \text{ha}^{-1}$ profile was too wet unless modelling could assume deeper incorporation than had been achieved in practice at the field site (Whelan et al., 2013). Thus, the results of these different studies all concur that, of the range of rates trialled, a single application of $500 \text{ t} \cdot \text{ha}^{-1}$ appeared to give the best overall performance in terms of physical soil improvement and future nutrient supply for plant growth.

The principal immediate or short-term gain from compost application is addition of SOM, from which then follows a range of related physical benefits, such as reduced bulk density, compaction and crusting, increased infiltration, porosity, water holding capacity and hydraulic conductivity (Cogger, 2005; Kilbride, 2014; Whelan et al., 2013). In addition, the chemical benefits include raising or stabilizing pH, the potential to immobilise metal contamination therein, reduce PTE mobility or plant uptake (Nason et al., 2007). Nutrient benefits include the addition of N and P, which are typically released slowly and without leaching of available forms (Claassen and Carey, 2004). Microbial activity is enhanced which helps release available P, especially for finer grades (Lata Verma and Marschner, 2013). If the compost is immature, woody or with a C/N ratio higher than 25:1, addition is likely to cause an initial reduction in available forms of both N and P (Bending et al., 1999; Claassen and Carey, 2007, 2004; Gagnon and Simard, 1999; Kilbride, 2014), although this can also be used to effect in mulching

with coarser grades for weed suppression (Sæbø and Ferrini, 2006). Other possible negatives include additional loadings of undesirable levels of the 7 PTEs (Cd, Cr, Cu, Pb, Hg, Ni, Zn) for which limits are specified (Anon, 2011), as shown in Table S1. For our trial these were sufficiently low to cause no regulatory concerns or significant issues in the soils, other than possibly for B (Lord, 2015). Both pot trials and field sites have shown that compost addition can release As into pore waters with dissolved organic carbon, especially when used as a mulch, with the latter then increasing the mobility of soil contaminants including As, Sb, Cr, Ni and Cd (Beesley and Dickinson, 2010; Clemente et al., 2010; Hartley et al., 2010). Macroscopic physical contaminants such as glass, metal and plastic are a major issue in the public acceptability of composts (Aspray, 2016; Echavarri-Bravo et al., 2017), even when derived from source-segregated organic wastes, which is now regarded as an emerging source of microplastic pollution in soils (Weithmann et al., 2018).

4.3. Long-term benefits and trade-offs of compost addition to soil

Our study has shown that the effects of a single application of compost during restoration can be detected in brownfield soil compositions even after 10 years and are still significantly different for high-rate applications of $500 \text{ t} \cdot \text{ha}^{-1}$ or $750 \text{ t} \cdot \text{ha}^{-1}$ for major nutrients, SOC and the key contaminants. For the majority of nutrients which remain in proportion to the original addition, the residual concentrations found correspond to an average annual decline of c 5% of the original, rising to roughly double this for more leachable (and so presumably more available) elements like K and B. Surprisingly, SOC also shows a similarly enhanced rate of decline at 7.4–9.5% per annum. This is in spite of the contrary increase in SOC found in the control area to which the amended areas have been compared. The increase found in the un-amended area (to 4.9%) compared to the baseline levels encountered 10 years earlier (1.8%) is presumably due to SOM accumulation in the upper layers of soil sampled here (0.15 m) as a result of plant growth from natural regeneration. Soil carbon averaging up to 9% was observed in naturally regenerated anthropogenic soils after 48 years (Salisbury et al., 2017). As these processes are also likely to have occurred in the amended and planted areas, and cannot be separated in our current analysis, the inference is that the rate of loss of C from the compost addition might be even higher than our estimate. However, the resultant net SOC increase remaining after compost addition in the upper 15 cm after 10 years still exceeds the equivalent of a 0.4% per annum absolute increase, compared to only 0.3% without treatment (Fig. 5). This is before any additional potential fixation of atmospheric CO_2 as inorganic carbon is accounted for, either by the natural pedogenic carbonation processes known to occur in compost-bearing technosols (Manning et al., 2013) or by reaction with calcium silicate or hydroxide in concrete demolition wastes in brownfield sites (Washbourne et al., 2015, 2012).

Literature information on the long-term behaviour of compost in brownfield soils, nutrient cycling and fate of its contaminants is limited, especially for field trials. For contaminants, studies have tended to focus on the composting process (Greenway and Song, 2002; Whittle and Dyson, 2002), utilisation issues (Hough et al., 2012), the implied immobilising benefits of compost addition (Smith, 2008) or observed leaching on application (Beesley and Dickinson, 2010; Hartley et al., 2010). For nutrients the maturity of compost affects the release rates (Al-Bataina et al., 2016) as well as the feedstock composition (Claassen and Carey, 2007). The reported rates of 1–3% N_T per annum, equivalent to an annual 10–50 kg available N per hectare, are lower than we observed. Although a first-order kinetic model can be applied to storm-event leaching, the results for multiple storm events were still found to be quasi-linear (Al-Bataina et al., 2016). Likewise, steady-state leaching was observed from extended incubation studies over a c. 2 year two years laboratory experiment (Claassen and Carey, 2004). The effects of compost mulch on pore water mobility of As and Sb in a brownfield soil were also seen to persist for at least 2 years (Clemente

et al., 2010). Significant effects on SOC, plant available P, K and Zn were found in agricultural soils 16 years after application of $50 \text{ t} \cdot \text{ha}^{-1}$ of composted dairy manure (Reeve et al., 2012), with yield increases indicating the half-life of the compost effects as 6 years. With the limited intervening data and sample variability it is difficult to determine whether our study shows linear or, more logically, an exponential decay. However, since the observed residual concentrations of N, P, Zn, Pb and Cu are approximately 50%, while those of SOC are 25%, the half-life effect of the compost appears to be around 10 years for these elements, compared to 5 years for SOC. Whether this increase in SOC can be considered to constitute mitigation of climate change depends on the alternative fate of the organic matter used to make the compost (Powelson et al., 2011). What is clear, however, is that compost application has extended benefits in subsequent years in brownfield soils for at least a decade and that both the initial leaching and long-term averaged annual release by mineralisation for application rates up to $500 \text{ t} \cdot \text{ha}^{-1}$ are unlikely to breach the Nitrates Directive limit of $170 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{a}^{-1}$ N (European Commission, 1991) without derogation, even before further plant or microbial reductions by assimilation or denitrification are discounted.

5. Conclusion

Previously developed “brownfield” sites offer an opportunity for dynamic carbon storage for at least a decade through the increase of SOC from addition of composted green waste to technosols created during landscaping or restoration activities. The relatively high rates of compost addition typically required for such a one-off application are acceptable without significant addition of anthropogenic contaminants or undesired rates of nutrient leaching to surface or groundwaters. Future work should be directed towards identifying and quantifying the various forms of SOC, anthropogenic and inorganic C in brownfields and similar non-agricultural landbanks.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2019.05.174>.

Acknowledgements

The British Land Reclamation Society is thanked for funding from the BLRS Legacy Investment Initiative that allowed the 10-year resampling of the trial site. Sunderland City Council are thanked for providing continued access to the site, which was based on pilots funded by the County Durham Environmental Trust. The Rainton Bridge field trial was originally established, monitored and evaluated with support from the EU Life III Environment Programme (LIFE05 ENV/UK/000125), the Waste and Resources Action Programme (ORG059, OBF001, OBF009, OBF010) and the Natural Environment Research Council (NE/H010440/1). Work on soil samples from the site by Amii Whelan was funded under the EPSRC Industrial Doctoral Training Partnership and OBF009 with co-supervision by Dr. Cedric Kechavarzi. Paula Lord is thanked for field assistance and Dr. Kate Walker for advice on statistics. Three anonymous reviewers are thanked for comments that greatly improved the final manuscript.

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Web links

- The “4 per 1000 initiative” <https://www.4p1000.org>.
- BioReGen life project. website. http://ec.europa.eu/environment/life/project/Projects/index.cfm?fuseaction=search.dspPage&n_proj_id=2833.
- <https://www.metoffice.gov.uk/public/weather/climate/gcwzefp2c>.
- www.nrm.uk.com/ Client area. Advice Sheet 38: Soil Organic Matter/Carbon – Which Method to Use? (Last accessed 25-7-18).